Laser produced highly ionized Aluminum plasma for high harmonic generation

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Introduction

- The purpose of this work is to devise highly ionized Al plasmas for HHG applications and to use them in future for increasing the energy cut-off of higher order harmonics
- In this work the aluminium plasma is generated using 1064 nm, 7 ns pulses, and the plasma is attuned spatio-temporally for Al IV emissions with irradiation energy as the control parameter, using time-resolved optical emission spectroscopy.
- An Al plasma plume rich in Al IV (Al 3^{+} ions) has a relatively large ionization potential of 120 eV.

High harmonic generation from gases

- The gas is irradiated by an intense laser field which generates free electrons by tunnel ionization
- The free electron is accelerated by the electric field of the laser and recombines with the parent ion, releasing XUVs or soft X-rays of sub-fs duration
- This process occurs if tunnel ionization dominates multiphoton ionization as determined from the value of the Keldysh parameter

- The Keldysh parameter

$$\gamma = \sqrt{I_p/2U_p}$$

Should be less than 1 (I_p and U_p are the ionization potential and the ponderomotive potential energy respectively).

- U_p is given by

$$U_P = e^2 E_0^2 / 4m\omega_0^2$$

where e and m are the electronic charge and mass, and E_0 and w_0 are the electric field amplitude and angular frequency of the driving laser pulse respectively.

HHG cut-off

- The energy cut-off for HHG is given (empirically) by

$$E_{Cutoff} \cong \hbar\omega_{max} = I_P + 3.17\ U_P$$

- Where ω_{max} is the maximum angular frequency. Since

$$U_P \propto I\lambda^2$$

Where I and λ are the intensity and central wavelength of the driving laser, energy cut-off can be increased either by increasing I_p , I and/or λ.

- According to the above equation, the typical harmonic order achieved is about 220 for an 800 nm laser pulse of intensity ≈ 10^{15} W/cm^2 in Helium
- An energy cut-off of 1.6 keV has been obtained from Helium, using 3.9 microns, 80 fs laser pulses

Some Limitations

- Even though the maximum photon energy of an HHG source is proportional to $I\lambda^2$, the photon flux is inversely proportional to λ^4. Therefore an optimum value of λ should be chosen for maximum HHG efficiency.

- This fact, in combination with the available laser technology, limits the drive wavelength to < 4 microns.

- Similarly, peak intensity cannot exceed a value I_{sat} which is proportional to ln I_p (although higher intensities are available from commercial laser systems).

- Finally, phase-mismatch arising from laser defocusing in the inhomogeneous high electron density LPP will reduce the HHG yield.

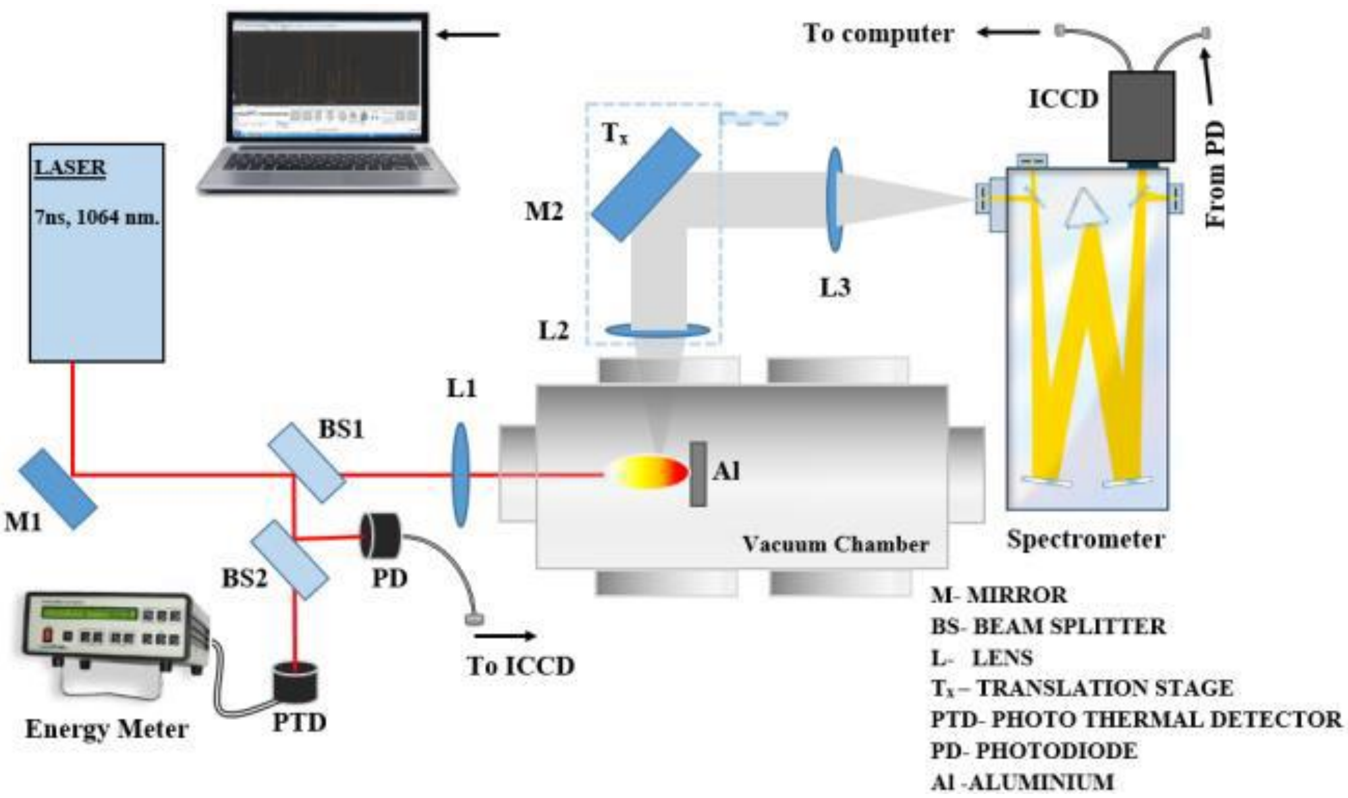
HHG from plasma media

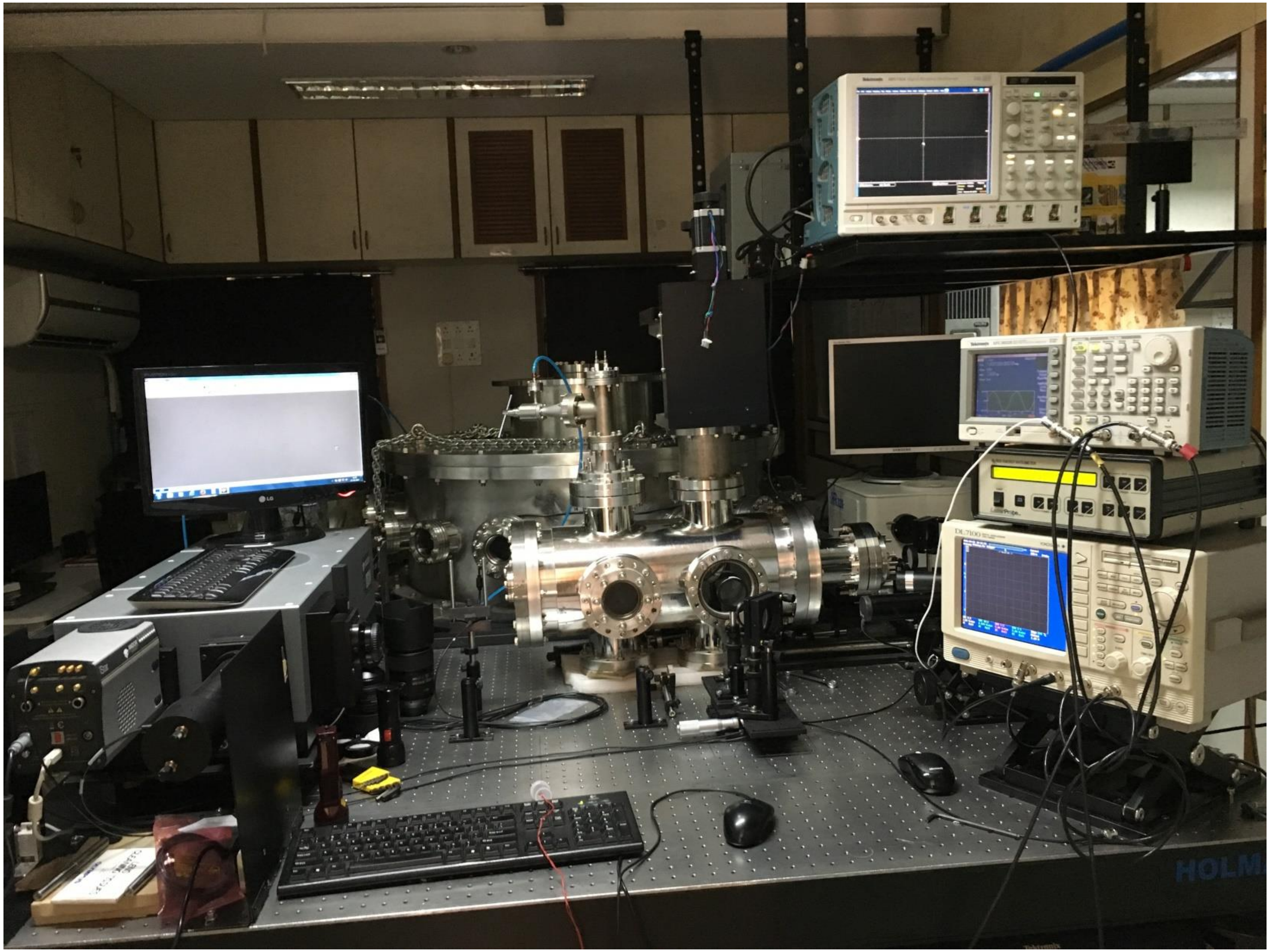
- Instead of the un-ionized (to start with) gas, a highly pre-ionized medium like the LPP (charge states of 3^+, 4^+ etc.) can be used as the medium for HHG generation

- Pre-ionization can be achieved using a high energy pump laser, into which the high intensity (ultrafast) pulse can be sent for generating the HHG. Laser defocusing is not a serious issue here

- This should yield high energy photons up to several keV due to the substantial increase in the ponderomotive energy of the free electrons

Experimental setup

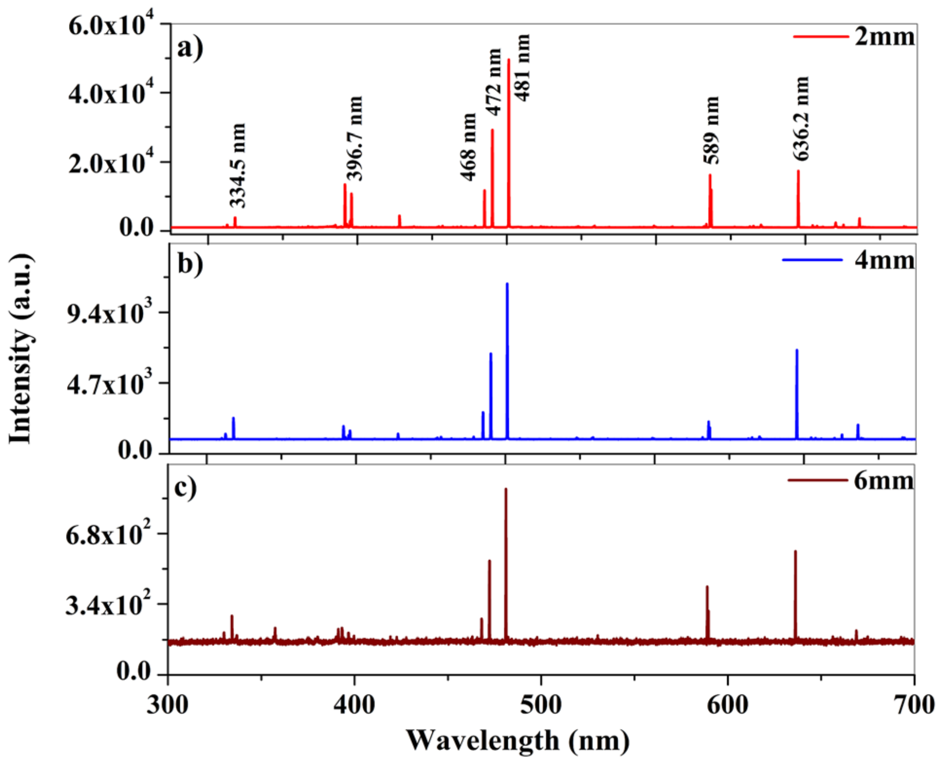



HOLM

Experimental parameters

- 99.99% pure Al target (from ACI Alloys Inc., USA); 50 mm x 50 mm x 3 mm size
- 1064 nm, 7 ns laser pulses
- Lens f = 500 mm, focal spot size = 400 microns
- Nitrogen background, 3 x 10^{-2} Torr
- 750 mm spectrometer equipped with a 13 x 13 micron, 1024 x 1024 Gen II ICCD (Andor), with resolution 0.02 nm.
- Transient dynamics of the species is measured by repeating the expt at different gate delays and gate widths of the ICCD
- Laser pulse energies used are 20 mJ to 85 mJ (16 J/cm^2 to 68 J/cm^2)

Electron temperature calculation



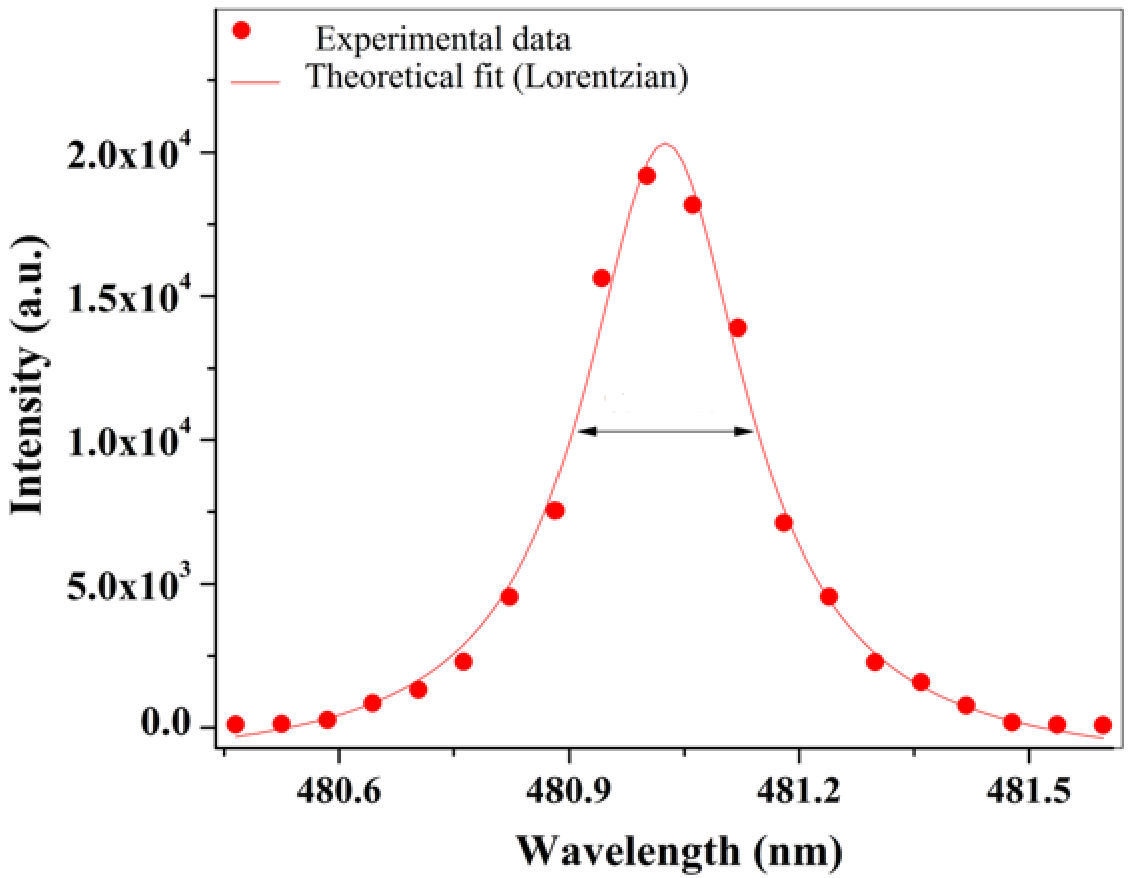
Line Ratio method:

$$\frac{I_1}{I_2} = \frac{g_1}{g_2}\frac{A_1}{A_2}\frac{\lambda_2}{\lambda_1}\exp\left[\frac{-(E_1 - E_2)}{K_B T_e}\right]$$

(Lower level should be the same for Both transitions)

I_i=line intensity, A_i=transition probability, g_i=statistical weight, λ_i=emission wavelength

Number density calculation from Stark broadening



Measurable Stark broadening occurs due to space charge effects. It is usually two orders of magnitude stronger than Doppler broadening.

(Doppler broadening is too small to be detected within the instrument resolution).

$$\Delta\lambda_{1/2} = 2\omega\left(\frac{N_e}{10^{16}}\right),$$

where ω is the electron impact parameter.

Al III and Al IV emissions

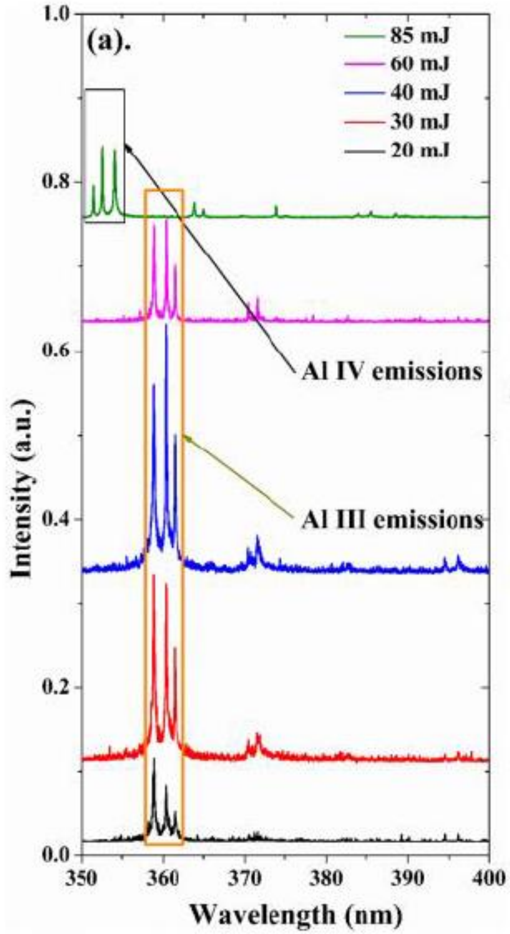
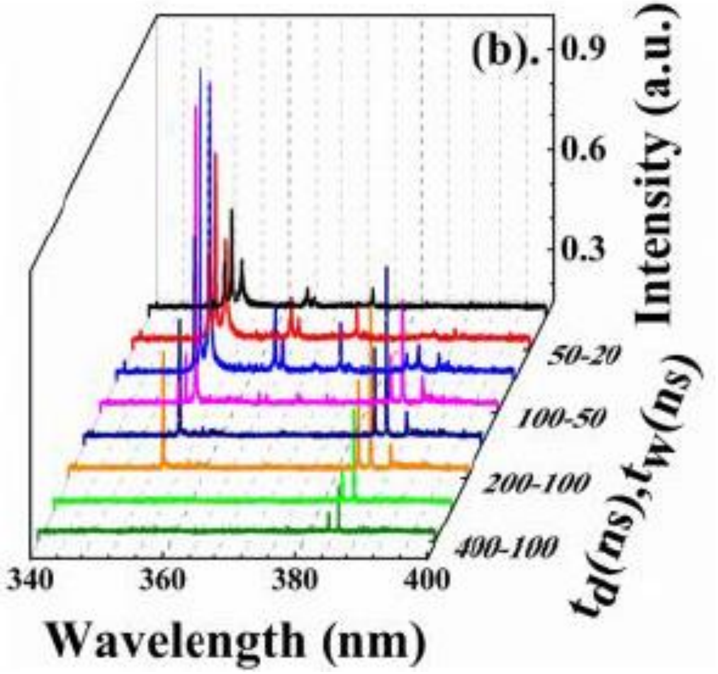


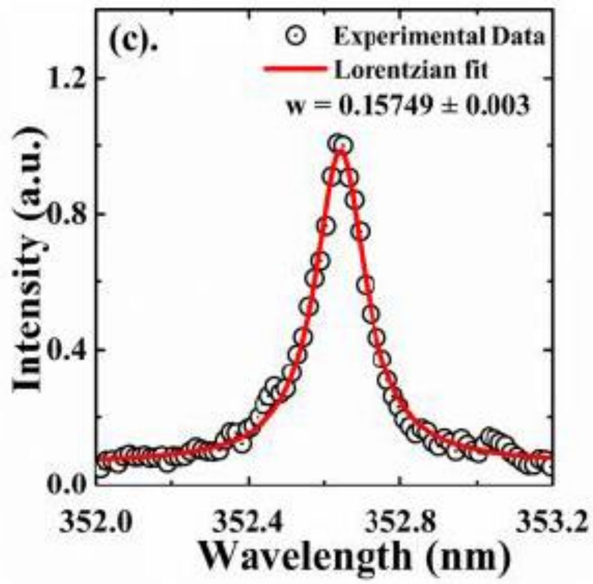
FIG 2. (a). Typical emission spectra measured for a gate width (t_w) of 50 ns and for a gate delay (t_d) 50 ns for various irradiation energies.

Emission Spectra



(b). Emission spectra measured for 85 mJ for various t_w and t_d. It is evident that the emission from Al IV maximizes for a time after irradiation (t_L) < 100 ns whereas recombination produces Al III and Al II for t_L > 100 ns.

Stark Broadening



(c). Lorentzian fit to the Al IV emission profile at 352.70 nm showing Stark broadening (FWHM = 0.16 nm). All measurements are done at a distance of 0.5 mm from the target surface. Background gas pressure is 3×10^{-2} Torr.

Intensity vs. Energy

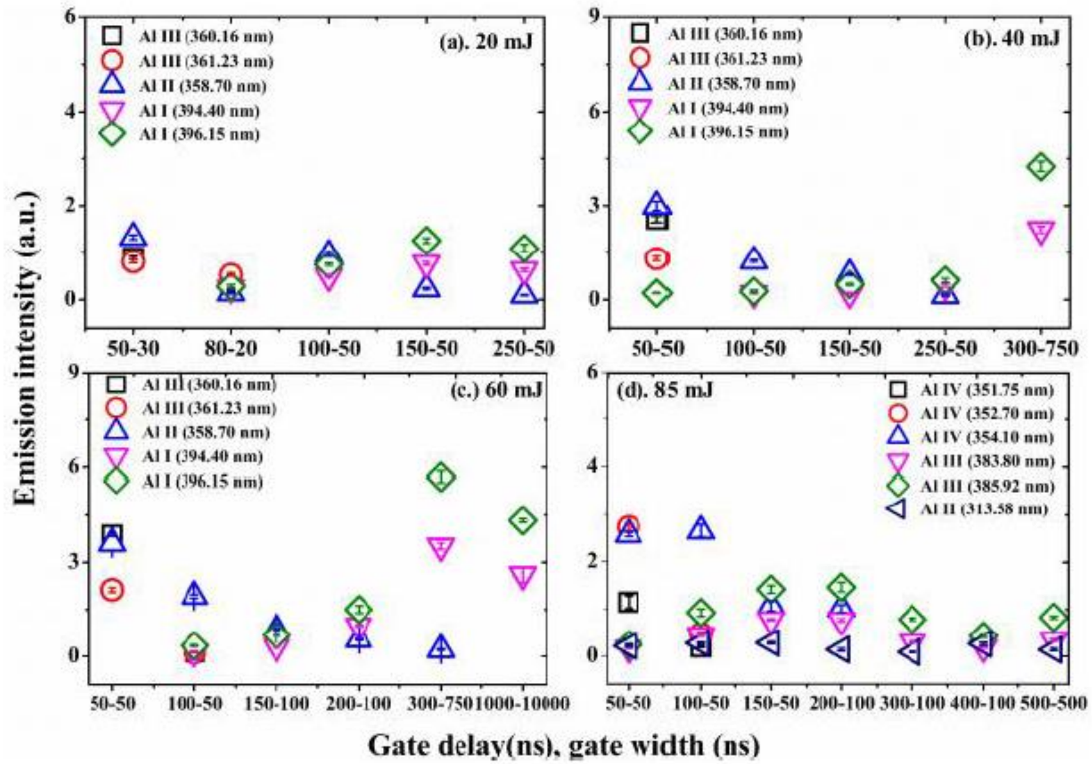


FIG 3. Emission intensity of Al I, Al II, Al III and Al IV spectral lines for different irradiation energies, measured at a distance of 0.5 mm from the target surface. Higher laser energies result in plasma with higher ionization states.

Line width vs. intensity

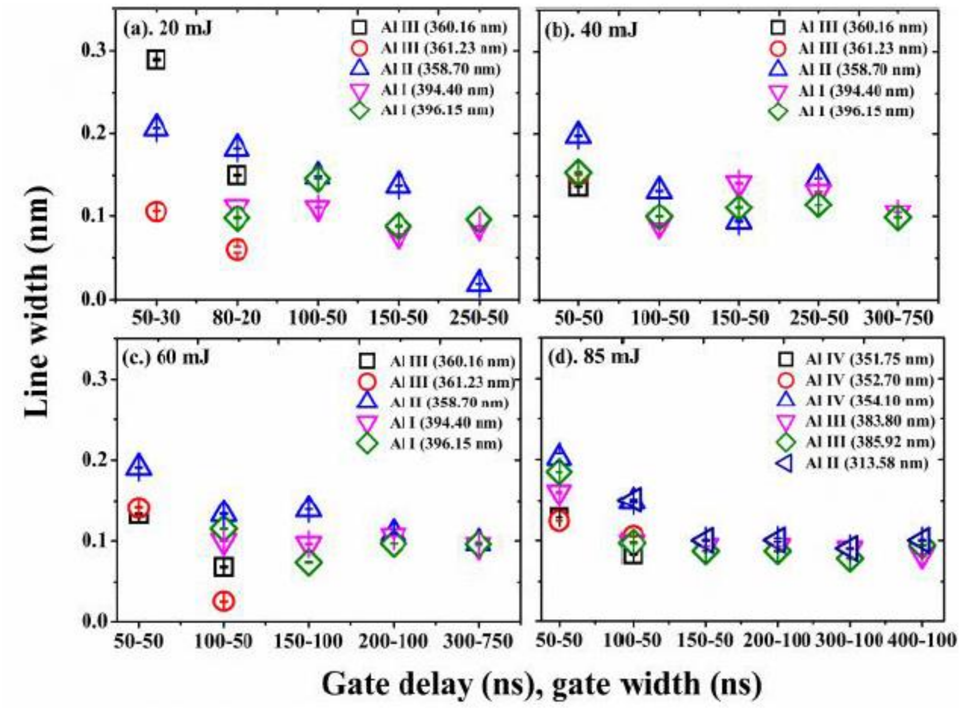


FIG 4. Estimated line width for ions and neutrals for different gate widths for various gate delays. It is clear from the figure that δλ reduces with expansion, i.e., as a function of time delay, this is due to the presence of large Coulomb forces at earlier times which decreases with time. It can also be inferred that δλ increases as a function of irradiation energy due to the presence of higher charged states and hence the large Coulomb field.

Stark width and emission intensity

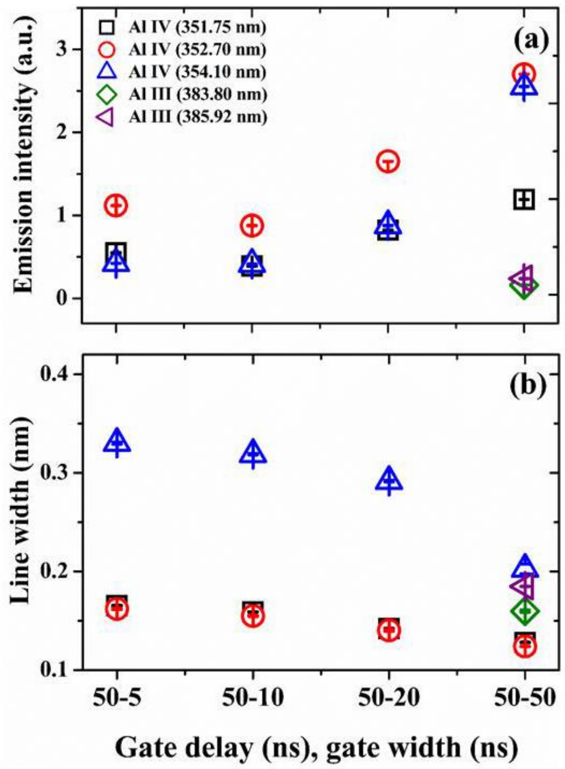


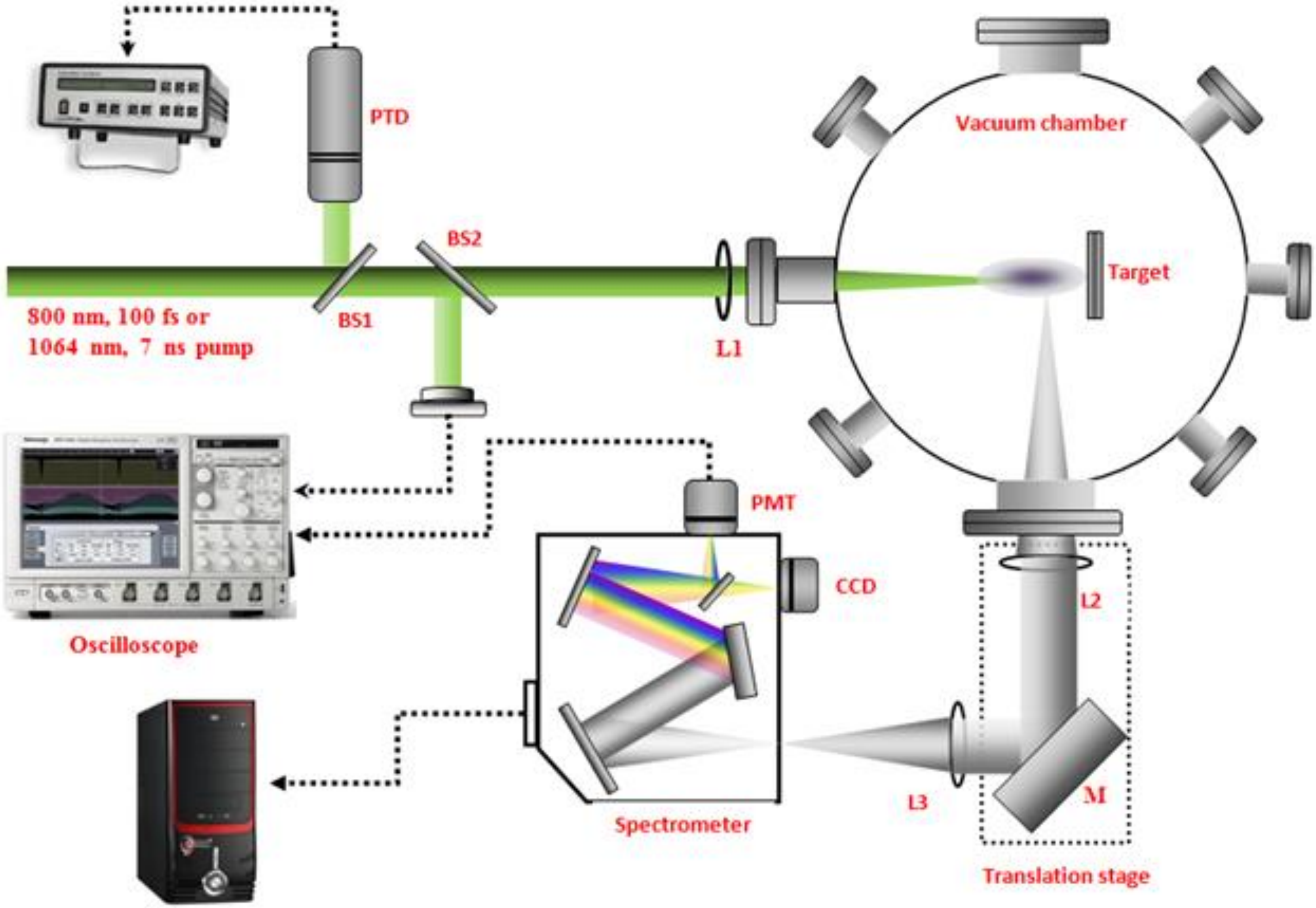
FIG 5. (a). Emission intensity and (b). Stark width measured for 85 mJ irradiation energy recorded for t_w< 100 ns for various t_w and t_d. It is clear that the plasma contains more Al IV emission in the initial time scales.

Summary

- Laser produced plasma have been generated in an Al target using 1064 nm, 7 ns laser pulses.
- At high irradiation energies emission from ionized species appear at earlier times in the spectra. A mix of ionized and neutral species is seen at later times.
- At the energy of 85 mJ, dominant Al IV emissions are seen close to the target (0.5 mm) at times < 70 ns (spatial and temporal optimization).
- The highly ionized Al plasma medium is expected to help raise the HHG cut-off to higher energies by irradiating the plasma at these earlier time scales with 800 nm, ≈ 30 fs pulses having irradiation intensity ≈ 6 x 10^{15} W/cm^2, once phase matching conditions are satisfied.

Thank You !

Experimental Setup



Spectral Lines

Table **I:** Line emission spectra from various species.

Species	Al **I**	Al **II**	Al **III**	Al **IV**
Wavelength (nm)	305.00	313.58	360.16	351.75
	308.21	331.55	361.23	352.70
	309.27	358.70	371.31	354.10
	394.40	365.10	383.17	363.88
	396.15	370.32	385.92	
		313.58		
		373.80		
		388.43		